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Determination of mercury in alcohol vinegar samples from Salvador, Bahia, Brazil

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ABSTRACT

This paper proposes a direct method for the determination of mercury in vinegar using ultrasound extraction and cold vapor atomic absorption spectrometry (CV AAS). The optimization step was performed using chemometric techniques, a full factorial design and a Box-Behnken matrix, where the factors studied were sodium tetrahydroborate flow rate, isoamyl alcohol volume, thiourea concentration and sonication time.

Using the optimized conditions, the method allowed the determination of mercury using external calibration, with limits of detection and quantification of 14 and 47 ng L^{-1}, respectively, and precision expressed as relative standard deviation of 3.87 and 8.11% for vinegar samples with mercury contents of 759.4 and 318.5 ng L^{-1}, respectively. There is no certified reference material of vinegar for evaluation of the accuracy; however, spike/recovery tests were performed in eleven samples. The recovery values ranged from 85 to 119%.

The method was used to determine the mercury content of eighteen alcohol vinegar samples acquired in Salvador City, Bahia, Brazil. The mercury content of these samples ranged from 51.9 to 759.4 ng L^{-1}.

1. Introduction

Vinegar is a condiment used worldwide in food preparation, which has been also employed as an antibacterial agent (Costa, Thomaz-Soccol, Paulino, & de Castro, 2009; Jafari, Falah-Tafti, Lotfi-Kamran, Zahraeii & Kazemi, 2012; Kuda, Yazaki, Takahashi, & Kimura, 2013, Parnell & Harris, 2003, Pereira, Rodrigues, & Ramalhosa, 2013; Sengun & Karapinar, 2005). Toxic elements such as lead, cadmium, mercury, etc. can occur in vinegar by contamination during production or storage. Given the importance of this ingredient in human food, analytical methods for quality control of vinegar are always very useful (Camin et al., 2013; Tsai & Kao, 2012; Ubeda, Balsera, Troncoso, Callejon, & Morales, 2012). Ndung'u, Hibdon, and Flegal (2004) evaluated sample preparation procedures during the determination of lead concentrations in vinegar by inductively coupled plasma mass spectrometry (ICP-MS) and graphite furnace atomic absorption spectrometry (GF AAS). Da Silva, Cadore, Nobrega, and Baccan (2007) proposed a direct method for determination of aluminum, barium, calcium, copper, potassium, magnesium, manganese and zinc in vinegar samples by axially viewed inductively coupled plasma optical emission spectrometry (ICP OES). The matrix interference was corrected using scandium as the internal standard. Dessuy et al. (2011) quantified cadmium and lead which had leached into vinegar from pewter cups by GF AAS using palladium–magnesium as a chemical modifier. Saei-Dehkordi, Fallah, and Ghafari (2012) determined lead, cadmium, copper and zinc in commercial Iranian vinegars using stripping chronopotentiometry. Fu, Nie, Xie, and Ferro (2013) developed a method for determination of 20 trace elements in vinegar by sector field inductively coupled plasma mass spectrometry (SF-ICP-MS). The sample decomposition procedure included microwave-assisted digestion. However, mercury was not determined in these studies. A published method for direct determination of mercury in vinegar was developed by Liu (2010), who used photochemical vapor generation in acetic acid media for sample introduction and atomic fluorescence spectrometry (AFS)

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as the detection technique. This method very fast, and has limit of quantification of 0.27 μg L^{-1}.

Mercury contamination is a matter of global concern because the main route of uptake is via food consumption (Cardoso, Afonso, Lourenco, & Nunes, 2013; Hernandez-Martinez & Navarro-Blasco, 2013). Therefore, improved procedures to quantify mercury in food samples are important (Da Silva, Portugal, Serra, Ferreira, & Cerda, 2013; Silva, da Silva, Leao, Matos, & Ferreira, 2012; Wang et al., 2013). The optimization of a method for determination of mercury in vinegar can be divided into two phases, i.e. "sample preparation" and "analytical measurement (quantification)". The quantification phase is very straightforward, using either CV AAS, cold vapor AFS (CV AFS) or ICP-MS, each of which has the necessary sensitivity for determination of mercury in these matrices. In contrast, the sample preparation phase includes several steps. The volatile character of mercury usually requires use of closed systems such as microwave assisted digestion with pressurized closed-vessel systems (Grindlay, Mora, Gras, & de Loos-Vollebregt, 2011), Parr digestion bombs (Karanasiou, Thomaidis, Eleftheriadis, & Siskos, 2005) and reflux systems using a cold finger (Ferreira et al., 2013; Oreste et al., 2013). The high content of organic compounds can be problematic, causing plasma instability and accumulation of carbon deposits on the cones and lens during ICP-MS analysis (Liu, 2010). For cold vapor generation coupled to AAS or AFS, a sample pre-treatment step also is required because some organic compounds may affect the mercury reaction with the tetrahydroborate (Capelo, Pedro, & Mota, 2003). Ultrasound extraction may be a good method for determination of mercury and other volatile elements in liquid and solid matrices. This technique would allow the direct determination of analyte, without heating or use of concentrated oxidizing acids. It also reduces the time required for analysis and minimizes the risks of contamination (Ferreira et al. 2010).

In this paper, a method for the determination of mercury in vinegar employing ultrasound extraction and cold vapor atomic absorption spectrometry (CV AAS) was tested. The sample preparation involved a sonication step using hydrochloric acid in the presence of thiourea. The optimization step was performed using a two level full factorial and Box-Behnken design, based on the work of Ferreira et al. (2007). The optimized method was then tested using eighteen alcohol vinegar samples from supermarkets in Salvador City, Brazil.

2. Material and methods

2.1. Instrumentation for the determination of mercury

Mercury was determined by High-Resolution Continuum Source (HR-CS) AAS using a CONTRAA 700 spectrometer (Analytik Jena, GLE, Berlin, Germany), with a high-intensity xenon short-arc lamp operated in the hot-spot mode, current of 13 A and wavelength of 253.6519 nm. The analytical signal was quantified by peak height. The vapor generator was operated manually using an HS50 hydride generator system (Analytik Jena, GLE, Berlin, Germany). The solutions were sonicated using a Model USC-1850 ultrasonic bath (UNIQUE, Indaiatuba, São Paulo, Brasil). Four milliliters of a 1% (w/v) sodium tetrahydroborate solution was injected (at a flow rate of 42 mL min^{-1}) into the reaction flask containing 10.0 mL of sample solution. Argon with a purity of 99.996% (White Martins, São Paulo, Brazil) was used as the carrier gas for the mercury vapor.

2.2. Reagents and samples

All reagents were prepared using high-purity water with resistivity of 18.2 mO, using water obtained from a Milli-Q Plus water purification system (Millipore, Bedford, MA, USA). Analytical-grade hydrochloric acid (Merck, Darmstadt, Germany) was double distilled in a duoPUR model 2.01E sub-boiling system (Milestone, Bergamo, Italy). Mercury calibration solutions at concentrations of 0.00; 0.50; 1.00; 1.50; 2.00; 2.50 and 3.00 μg L^{-1} were prepared daily by the serial dilution of a 1000 mg L^{-1} stock solution (Merck, Germany) with 0.05% (v/v) nitric acid. Spike experiments were performed with additions of mercury concentrations of 100 and 500 ng L^{-1}. The reductant was 1% (w/v) sodium tetrahydroborate, stabilized with 0.05% (w/v) sodium hydroxide. It was prepared daily using analytical grade reagents (Merck, Germany) and was filtered through a 0.45-μm membrane (Millipore, Brazil). The 2.0% (w/v) thiourea solution used as a chelating agent was prepared by dilution of the reagent (Merck, Germany) with high-purity water.

The samples analyzed were alcohol vinegars with acetic acid content in the range from 4 to 6%, produced by the fermentation of ethanol by acetic acid bacteria. Other constituents included: ethanol (0.7–2.2 g L^{-1}), methanol (0.1–11.8 mg L^{-1}), ethyl acetate (112–265 mg L^{-1}), some metals (with content at mg L^{-1} levels) and other organic compounds.

2.3. Sample preparation and the determination of mercury by CV AAS

A 12.5 mL vinegar sample was added to a 25 mL volumetric flask, together with 3.10 mL of concentrated hydrochloric acid and 5 mL of the 2% (w/v) thiourea solution. The flask was placed in a 50 °C ultrasonic bath for 20 min. After cooling to room temperature (25 °C), ultrapure water was added to a total volume of 25 mL, a 10 mL aliquot was transferred to the 50 mL reaction flask of the hydride generator, and 400 μL of isoamyl alcohol was added as an antifoaming agent. Sodium tetrahydroborate (4 mL) was added to the reaction flask and the vapor that was generated was carried into a quartz T-tube which was coupled to the AAS spectrometer. The blank reagent was prepared in the same manner except without a vinegar sample. The volumes and concentrations of sodium tetrahydroborate and hydrochloric acid chosen for this analysis were based on the optimal conditions reported by Silva et al. (2012).

2.4. Multivariate optimization of the experimental factors

Chemometric tools such as two-level full factorial and Box-Behnken designs have often been employed for optimization of processes and as analytical methods (Alves & Coelho, 2013; Bevilacqua, Campaniello, Speranza, Sinigaglia, & Corbo, 2013; Escudero, Cerutti, Martinez, Salonia, & Gasquez, 2013, Reboredo, Gonzalez-Barreiro, Cancho-Grande & Simal-Gandara, 2012). Advantages over the univariate methodology are reduction of the number of experiments necessary and the opportunity to evaluate the interactions between the factors in the chemometric techniques. The effect of a factor was calculated as the difference between the average response at the highest level for this factor and the average response at its lowest level. The positive or negative signs and numerical values obtained are used for evaluation of the effects of factors on the studied system (Massart et al., 1997).

Optimization of the method was developed in two steps. First, a two-level full factorial experiment was carried out using four factors (sodium tetrahydroborate flow rate, isoamyl alcohol volume, thiourea concentration and sonication time) and the data were assessed using a Pareto chart. The central points were analyzed in triplicate to determine the experimental error and also to provide data for a curvature test (Massart et al., 1997). The second step was analysis using a Box Behnken design to determine the critical conditions of the principal factors. Absorbance (measured as peak height) was the measured chemometric response.

The experiments were performed in random order. The data were evaluated using the Statistica 6.0 statistics program (Statistica for Windows, 1999).

3. Results and discussion

3.1. Optimization of the experimental conditions for vapor generation of mercury

The raw results of the full factorial design involving the experimental variables: sodium tetrahydroborate flow rate, isoamyl alcohol volume, thiourea concentration and sonication time are shown in Table 1.

These data were analyzed statistically to determine the effects of the factors and their interactions. All four main effects were significant. The effect of thiourea concentration was positive (effect: 0.0088 ± 0.0014). Thiourea acts as a chelating agent, improving the reduction reaction, likely by aiding the decomposition of the complexes of mercury ions and organic acids in vinegar. The mean quantity of mercury released from the analytical matrix, as indicated by absorbance values in Table 1, was considerably higher in the presence of thiourea than in its absence (absorbance peaks 0.01783 and 0.00901, respectively). Sodium tetrahydroborate flow rate also had a positive effect as shown by the absorbance values (effect: 0.0078 ± 0.0014). This suggests an increase in the rate of the reduction reaction at the higher flow rate. Isoamyl alcohol was tested as an antifoaming agent. This had also a positive effect (effect: 0.0018 ± 0.0014), which suggests that increasing this factor increased the mercury vapor generation, most likely because the isoamyl alcohol facilitated the liberation of the reduced elemental mercury from the liquid to the gaseous phase. Sonication time had a very small effect on vapor generation (effect: −0.0015 ± 0.0014). This means that mercury was lost, during the sample preparation when the longer sonication time was used. This loss was probably in elemental form by volatilization (Welz & Sperling, 1999). All the interactions with thiourea concentration were significant. This highlights the importance of the chelation reaction between mercury(II) ions and thiourea, which improved mercury extraction in the sample preparation step for the generation of cold vapor (Aceto, Foglizzo, Mentasti, Sacchero, & Sarzanini, 1995). The negative sign for the interaction between thiourea concentration and sonication time (effect: −0.0046 ± 0.0014) indicates that the best conditions for sample preparation are the higher concentration of thiourea (0.10%) with the lower sonication time (20 min). The interaction between thiourea concentration and isoamyl alcohol volume was positive (effect: 0.0030 ± 0.0014) indicating that the higher thiourea concentration was more effective when the higher volume of isoamyl alcohol was also present. The interaction between thiourea concentration and sodium tetrahydroborate flow rate was positive (effect: 0.0032 ± 0.0014). The reduction reaction using sodium tetrahydroborate is fast and the analytical signals measured during the experiments are based on peak height, which explains why the increased thiourea concentration and sodium tetrahydroborate flow rate increased the observed measurement, indicating a positive interaction between these two factors. The interactions: (isoamyl alcohol volume × sonication time), (isoamyl alcohol volume × sodium tetrahydroborate flow rate) and (sodium tetrahydroborate flow rate × isoamyl alcohol volume) were not significant.

A curvature test was applied to the experimental data, using the equation: Curvature = RFD − RCP, where RFD is the average of the responses obtained from the factorial design, and RCP is the average of the responses from the central points, i.e. RFD = 0.01342, RCP = 0.01909 and Curvature = −0.00567. The negative value resulting from this calculation indicates that the maximum vapor generation occurs in the region of the central point of the factorial design (Massart et al., 1997).

Based on these results, a Box-Behnken design was used to determine the optimal conditions of thiourea concentration, sonication time and sodium tetrahydroborate flow rate, having absorbance (peak height) as the measured response. The isoamyl alcohol volume was fixed at 400 μL because this amount prevents formation of any bubbles during vapor generation. The experimental design (coded and real values) and the responses are shown in Table 2.

Statistical analysis of the results fitted a quadratic model, and suggest the following absorbance maxima for thiourea concentration: 0.41%; sonication time: 19.79 and sodium tetrahydroborate flow rate: 41.76 mL min^{-1}. Combining these results with previous work by Silva et al. (2012), the optimal conditions for the determination of mercury were identified as hydrochloric acid concentration: 1.5 mol L^{-1}; sodium tetrahydroborate concentration: 1.0%;

Table 1
Examination of experimental conditions for determination of mercury in vinegar using two-level full factorial design.

Exp.	NaB-FR (mL min^{-1})	Alcohol (μL)	Thiourea (%)	Time (min)	Abs (peak height)
1	+ (30)	+ (400)	+ (0.1)	+ (40)	0.02111
2	+ (30)	+ (400)	+ (0.1)	− (20)	0.03318
3	+ (30)	+ (400)	− (0.0)	+ (40)	0.01209
4	+ (30)	+ (400)	− (0.0)	− (20)	0.00795
5	+ (30)	− (200)	+ (0.1)	+ (40)	0.01457
6	+ (30)	− (200)	+ (0.1)	− (20)	0.02448
7	+ (30)	− (200)	− (0.0)	+ (40)	0.01690
8	+ (30)	− (200)	− (0.0)	− (20)	0.00831
9	− (14)	+ (400)	+ (0.1)	+ (40)	0.01248
10	− (14)	+ (400)	+ (0.1)	− (20)	0.01460
11	− (14)	+ (400)	− (0.0)	+ (40)	0.00629
12	− (14)	+ (400)	− (0.0)	− (20)	0.00707
13	− (14)	− (200)	+ (0.1)	+ (40)	0.01098
14	− (14)	− (200)	+ (0.1)	− (20)	0.01120
15	− (14)	− (200)	− (0.0)	+ (40)	0.00688
16	− (14)	− (200)	− (0.0)	− (20)	0.00658
CP	0 (14)	0 (300)	0 (0.05)	0 (30)	0.01922
CP	0 (14)	0 (300)	0 (0.05)	0 (30)	0.01836
CP	0 (14)	0 (300)	0 (0.05)	0 (30)	0.01970

NaB-FR – sodium tetrahydroborate flow rate, Alcohol – isoamyl alcohol volume, Thiourea – thiourea concentration, Time – sonication time, Abs – absorbance, CP – central point.

Table 2
Optimization of the experimental conditions for determination of mercury in vinegar using Box Behnken design.

Exp.	Thiourea (%)	Time (min)	NaB-FR (mL min^{-1})	Abs (peak height)
1	−1 (0.2)	−1 (5)	0 (35)	0.02283
2	1 (0.6)	−1 (5)	0 (35)	0.03897
3	−1 (0.2)	1 (25)	0 (35)	0.04258
4	1 (0.6)	1 (25)	0 (35)	0.03677
5	−1 (0.2)	0 (15)	−1 (22)	0.02160
6	1 (0.6)	0 (15)	−1 (22)	0.01681
7	−1 (0.2)	0 (15)	1 (48)	0.03543
8	1 (0.6)	0 (15)	1 (48)	0.04846
9	0 (0.4)	−1 (5)	−1 (22)	0.03046
10	0 (0.4)	1 (25)	−1 (22)	0.03535
11	0 (0.4)	−1 (5)	1 (48)	0.04306
12	0 (0.4)	1 (25)	1 (48)	0.06946
CP	0 (0.4)	0 (15)	0 (35)	0.06730
CP	0 (0.4)	0 (15)	0 (35)	0.06415
CP	0 (0.4)	0 (15)	0 (35)	0.07062

Thiourea – thiourea concentration, Time – sonication time, NaB-FR – sodium tetrahydroborate flow rate, Abs – absorbance, CP – central point.
The numbers in parentheses which follow the Box Behnken codes are the actual quantities used in the experiments.

sodium tetrahydroborate volume: 4.0 mL; thiourea concentration: 0.41%; sonication time: 19.8 min; sodium tetrahydroborate flow rate: 42.0 mL min^{-1} and isoamyl alcohol volume: 400 μL.

3.2. Method validation

Since vinegar is a complex matrix, the addition analyte (spike) method was employed to determine the calibration technique that should be used. A vinegar sample (with low mercury content) was spiked with mercury(II) ion in the concentration range of 0.0–3.0 μg L^{-1}, to produce a calibration curve. Another calibration curve was prepared using 1.5 M hydrochloric acid solution instead of vinegar, and again the concentration of the mercury spike was varied from 0.0 to 3.0 μg L^{-1}. The slopes of the two calibration curves (expressed at the 95% confidence level) were 0.0664 ± 0.0020 and 0.0677 ± 0.0021 for vinegar and hydrochloric acid, respectively. The similarity of these results demonstrated that the external calibration technique can be used to determine the concentration of mercury in vinegar samples using standards prepared in 1.5 M hydrochloric acid. The analytical limits were calculated as limit of detection (LOD) = (3 s/S) and limit of quantitation (LOQ) = (10 s/S), where s is the standard deviation of a blank and S is the slope of the calibration curve (Massart et al., 1997). Under the optimized conditions, LOD and LOQ were 14 and 47 ng L^{-1}, respectively.

Using hydrochloric acid and thiourea improved the mercury extraction and increased the sensitivity of the method, reflected in the lower limits of detection and quantification compared with those reported by Liu (2010). The precision was calculated as relative standard deviation, i.e. 3.87 and 8.11% for samples with mercury contents of 759.4 and 318.5 ng L^{-1}, respectively.

3.3. Application – determination of mercury in vinegar samples

The proposed method was applied for determination of mercury in eighteen alcohol vinegar samples acquired in local supermarkets (Table 3). The mercury content measured in these samples varied from 51.9 to 759.4 ng L^{-1}, which is consistent with results reported by Liu (2010). There is no certified reference material of vinegar. However, spike/recovery tests carried out on eleven of the 18 vinegar samples had recovery values of 85–119%. All samples were analyzed in triplicate.

Table 3
Mercury content of alcohol vinegar samples purchased in Salvador City, Brazil.

Sample	Hg spike added (ng L^{-1})	Hg content (ng L^{-1})	Spike recovery (%)
1	0	51.9 ± 14.9	0
	100	140.6 ± 33.9	89
2	0	122.7 ± 26.8	–
	100	229.1 ± 25.7	106
3	0	70.1 ± 11.2	–
	100	163.0 ± 23.1	93
4	0	102.4 ± 33.8	–
	100	205.6 ± 37.9	103
5	0	164.8 ± 31.3	–
	500	756.0 ± 35.4	118
6	0	103.7 ± 31.2	–
	500	614.6 ± 12.1	102
7	0	100.3 ± 1.8	–
	500	614.6 ± 12.1	114
8	0	106.5 ± 20.7	–
	500	531.4 ± 28.8	85
9	0	114.2 ± 42.1	–
	500	710.9 ± 45.4	119
10	0	286.7 ± 24.0	–
	500	761.7 ± 50.3	95
11	0	318.5 ± 41.1	–
	500	868.50 ± 63.1	110
12	0	567.3 ± 16.2	–
13	0	486.5 ± 10.5	–
14	0	206.9 ± 32.5	–
15	0	280.4 ± 61.8	–
16	0	759.4 ± 46.7	–
17	0	58.1 ± 25.6	–
18	0	567.3 ± 16.2	–

4. Conclusions

Vinegar is a complex matrix, composed of several organic compounds. However, the experimental conditions identified in this method allow the direct determination of mercury using external calibration, with good precision, accuracy and sensitivity. Use of thiourea as a chelating agent and sonication for 20 min enabled a simple sample preparation. The mercury contents determined in this work are in agreement with those reported in the literature.

The method is likely to be important in view of the high toxicity of this element combined with the large consumption of this condiment in the human diet. These results are important because this type of vinegar (i.e. alcohol vinegar) is consumed daily by most Brazilians.

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